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AUTHOR(S): j. R. Travis

B. D. Nichols

J. W. Spore

T. L. Wilson

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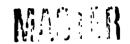
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LOS Alamos National Laboratory
Los A'amos, New Mexico 87545







AN HMS/TRAC ANALYSIS OF A HIGH-LEVEL RADIOACTIVE WASTE TANK

J. R. Travis
Science Applications International Corporation

B. D. Nichols
J. W. Spore
T. L. Wilson
Los Alamos National Laboratory

ABSTRACT

It has been observed that a high-level radioactive waste tank generates quantities of hydrogen and nitrous oxide mixtures that are potentially well within flammability limits. These gases are produced from chemical and nuclear decay reactions in a slurry of radioactive waste material. The slurry is covered by a thick crust composed of sodium nitrate and nitrite salts. Significant amounts of the combustible and reactant gases are produced over a 3- to 4-month period before the crust ruptures and the gases are vented into the air cover gas space above the crust. Postulating an ignition of the hydrogen/nitrous oxide/air mixture after this venting into the cover gas, we nave calculated the pressure and temperature loading on the double-walled waste tank with the three-dimensional, time-dependent fluid dynamics coupled with chemical kinetics HMS (Hydrogen Mixing Studies) computer code. The waste tank has a ventilation system designed to maintain a slight negative gage pressure during steady-state operation. We have modeled the ventilation system with TRAC (the Transient Reactor Analysis Code), and we have coupled these two best-estimate accident analysis tools to provide the ventilation response to pressures and temperatures generated by the hydrogen burn. Significant pressures are produced by this event, and the threat to the tank's integrity currently is being evaluated.

I. INTRODUCTION

The Hanford Site, which is located on the Columbia River in southeastern Washington state, was established in 1943 by the federal government primarily for plutonium production for national defense purposes. Inherent in the processing of spent reactor fuels for plutonium production is the production of radioactive waste. In recent years, waste management has become the central focus of activity at the site.

The bulk of these radioactive liquid wastes is stored underground at the Hanford Tank Farm. The Tank Farm comprises 149 single-shell tanks and 28 double-shell tanks. The single-shell tanks range in capacity from 208.2 to 3785.0 m³ (55,000 to 1.0 million gal); the double-shell tanks have a capacity of 4163.5 m³ (1.1 million gal). The single-shell tanks are constructed of a carbon-steel liner surrounded by reinforced concrete.

The double-shelled tanks are constructed of a carbon-steel tank within a carbon-steel liner surrounded by reinforced concrete. Figure 1 shows a schematic view of the SY farm. Each of the double-walled tanks has an inner shell diameter of 22.86 m and an outer shell diameter of 24.384 m. The outer shell is encased by a 0.4572-m-thick reinforced concrete tank to give a total diameter of 25.2984 m. The intended purpose of the double-shell tanks was to receive stabilized and concentrated waste from the single-shell tanks.

The concentrated slurry was first produced and stored in Tank 101-SY in 1977. This first batch of slurry exhibited a steady and unexplained growth in volume at a rate of about 1%/month. Since the last deposit of concentrated slurry in this tank in 1980, the waste surface has been rising and abruptly falling cyclically approximately once every 3 or 4 months. A series of parametric studies showed that various gases were generated as the organics degraded, including hydrogen, nitrogen, nitrous oxide, and carbon dioxide. These studies also indicated that the gas bubbles absorb into fine solids in the waste, causing the solids to float upward. This process continually adds to the thickness of a solid layer of material on the surface of the waste that forms a crust. It was postulated that as gas accumulates under the crust, the crust itself gradually rises within the tank, until increased gas volume and pressure lead to a break or fissure in the crust layer and the release or "burp" of the under-crust gases. Indeed, measured vapor space pressurization accompanied by waste level subsidence has occurred regularly in Tank 101-SY. During these over-pressurization events, the concentrations of hydrogen approached and occasionally exceeded the threshold of hydrogen flammability in dry air.

In 1990, the US Department of Energy (DOE) Office of Environmental Restoration and Waste Management (EM) created a Task Force to address these safety concerns.[1] One of the problems the Task Force was to focus its efforts on was the flammable gas (hydrogen) accumulation issues related to tanks at the Hanford Site. This Task Force requested that a detailed reanalysis of a postulated hydrogen accident be done. The work presented in this paper is a result of the response of the Engineering and Safety Analysis Group (N-6) of the Los Alamos National Laboratory to this request.

II. LOS ALAMOS APPROACH

The Los Alamos goals were to re-examine the hydrogen issues and accurately model the phenomenology of hydrogen behavior in the SY Tank Farm. To accomplish this task, it was necessary to investigate hydrogen behavior in three-dimensional volumes connected with a ventilation system. The dynamics of a hydrogen release required modeling low-speed, buoyancy-driven flows with the possibility of combustion occurring in the release tank and perhaps propagating through the ventilation system.

The analytical tools used to model these phenomena were the coupled HMS [2] (Hydrogen Mixing Studies) and TRAC [3] (Transient Reactor Analysis Code) computer codes. HMS is a finite-volume computer code that solves the transient, three-dimensional, compressible Navier-Stokes equations with multiple species coupled with chemical kinetics. It was designed at Los Alamos to be a host-estimate tool for predicting the transport, mixing, and combustion of hydrogen gas in nuclear reactor containments. It was used in this study to model the release of hydrogen and nitrous oxide into the vapor space of Tank 101-SY. These released gases then were transported and mixed in the cover gas volume according to local

dynamics (such as convection and turbulent diffusion) before an ignition point was postulated. After ignition of the hydrogen, the flows were driven by the coupled fluid-dynamics/chemical kinetics algorithm.

TRAC is a finite-volume thermal-hydraulics code developed at Los Alamos for advanced best-estimate predictions of postulated accidents in light-water-reactors. The network flow capability of TRAC was used to model the ventilation system associated with Tank Farm SY. In this context, Tanks 102 and 103 were modeled using the lumped volume capability, and the ventilation system, including the in-leakage ports to each tank, were modeled with the one-dimensional finite-volume capability of TRAC.

The two codes were coupled numerically at the physical boundary of Tank 101. That is, HMS provided TRAC pressures, temperatures, and gas composition at computational cells adjacent to the physical connections for the ventilation system and the in-leakage port. TRAC used these values to compute flow rates throughout the entire system excluding Tank 101. The resulting TRAC velocities representing the response of the ventilation system then were used as inflow or outflow boundary conditions for HMS. This coupling was accomplished in a simultaneous explicit manner that proved to be extremely stable and robust.

III. HMS/TRAC WASTE TANK ANALYSIS

Although there are several opinions on the composition of the under-crust gas mixture, we present here what is considered a worst case. We assume that the gas is made up of a homogeneous 50% hydrogen/50% nitrous-oxide mixture based on a volume bases. The total mass released to the tank dome is 230 kg in 255 m³, thus breaking down into 10 kg of hydrogen and 220 kg of nitrous oxide in 127.5 m³, respectively. This source is assumed to enter the tank dome volume during a 5-min release period. The injection flow distribution starts from a zero flow rate followed by a linear ramp to its maximum value over 100 s. For the next 100 s, there is a constant release equal to the maximum of the ramp up, and the final 100 s is an exponential decay with a 50-s half-life. At the end of this injection, which is assumed to occur at a maximum distance from the ventilation system penetration into the tank, an energy source ignites the mixture at the release location. A deflagration moves radially out from this ignition point, consuming most of the hydrogen in less than 1 s. As a result of the hydrogen combustion in the nitrous-oxide/air environment, the tank is loaded to a maximum pressure of 475 kPa and an average temperature of 1510 K.

During steady-state operation, the ventilation system fan holds the tank pressure at about a negative 1 kPa (10 cm water) relative to the outside pressure. This ensures that tank cover gas volume of 935 m³ has a turnover rate of about 1 h because the air exchange is roughly 16.5 m³/min. Because of the ventilation and in-leakage flow areas, this relates to flow velocities of 3.8 m/s and -9.0 m/s, respectively, to the ventilation system and in-leakage ports. Note that a positive velocity is out of the tank into the ventilation system and a negative velocity is into the tank.

During the release phase, a slightly greater than atmospheric pressure of about 1.2 kPa (13 cm water) is achieved in the tank dome during the constant maximum release time between 100 and 200 s. This causes a flow reversal in the in-leakage port (attaining about 12-m/s outflow) and an increase in the flow out of the tank's ventilation system connection (to almost 14 m/s). However, by the end of the 300-s release, because of the exponential decay behavior, the

tank pressure and flow velocities returned to near-normal operating conditions. Because of this transient and the mixing phenomena within the tank, approximately 13% of the injected hydrogen and nitrous oxide has vented from the tank. This leaves roughly 8.7 kg of hydrogen, 191 kg of nitrous oxide, 238 kg of oxygen, and 785 kg of nitrogen in the tank at ignition. The mixture is not well mixed in the time scale of 5 min, and because the injected gases are released at a higher temperature than the tank cover gas temperature, the burp tends to create a plume that rises to the tank top, causing a slightly enriched region of fuel above the mid-height at ignition.

After the injection phase, we consider five gaseous components in the tank and ventilation system: H_2 , N_2O , O_2 , N_2 , and H_2O . We model the global chemical kinetics reactions as

(1)
$$H_2 + 1/2 O_2 \rightarrow H_2O + 57.8 \text{ kcal/mole } H_2 \text{ consumed and}$$

(2)
$$H_2 + N_2O -> H_2O + N_2 + 77.4 \text{ kcal/mole } H_2 \text{ consumed.}$$

Not only is the nitrous oxide a more aggressive reactant, it is a more energetic reactant as well, releasing about one-third more energy than the hydrogen-oxygen reaction. We model these reactions using one-step finite rate chemistry, where the Arrenihus reaction rate is weighted according to the volume fractions of available oxidizer needed for (1) and (2) as applied to every computational control volume.

We show the results of the HMS/TRAC calculation in Figs. 2–5. The time histories of all gaseous components showing the mass inventory are presented in Figs. 2–4. Only the last few seconds of the burp phase are shown with ignition taking place between 300 and 301 s. Over the next second, most of the hydrogen is burned and the tank pressure rises to its maximum as seen in Fig. 5. As also shown in Fig. 5, the average tank temperature reaches its maximum within a few seconds as the last amounts of hydrogen are consumed. Within a few seconds of ignition, the depressurization of the tank begins. When longer term pressure histories are examined, it appears that the tank depressurizes with about a 25-s decay constant, so the tank pressure has nearly returned to normal conditions after 100 s.

IV. CONCLUSIONS

There is no question that the filters are no longer effective after this event, and these loads could cause the tank to fail. Preliminary structural analyses indicate that when exposed to the dynamic pressure loads resulting from a worst-case gas release as described above, the inner shell could fail near the bottom and possibly the outer shell as well. Some novel solutions to this problem must be developed. It is important to isolate Tank 101 from the other tanks in the SY farm. The advantages are that if such an energetic event occurs, Tanks 102 and 103 remain on an operating ventilation system. Isolation of Tank 101 with its own ventilation system would allow upgrading the performance to permit an increase of air exchanges with the outside air. This increases the potential mixing during and following the release phase when the gas mixture is most sensitive.

Another modification that would enhance the volumetric mixing during the most critical time would be to redesign the in-leakage port. The idea would be to direct the in-leakage

flows in such a manner that the entire tank dome volume participates in the convective flows. Designing a flow sparger and installing it as a extension to the in-leakage port could help.

It might be possible to introduce a fire suppressant such as Halon (or possibly a more environmentally acceptable compound) into the tank during a release to prevent a combustion event. This option could be very expensive because of the cost of the chemicals and the design may call for recovering the fire suppressant rather than allowing it to vent to the atmosphere.

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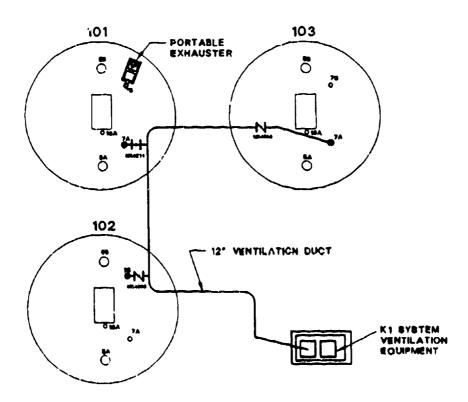


Fig. 1. Existing SY Farm ventilation system.

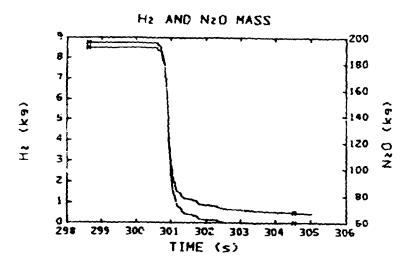


Fig. 2. Time history of H₂ and N₂O.

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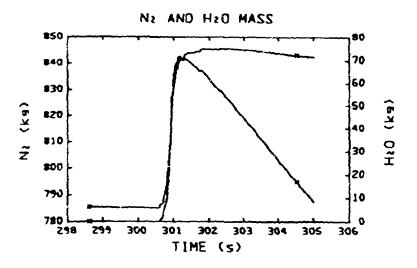


Fig. 4. Time history of N2 and H20.

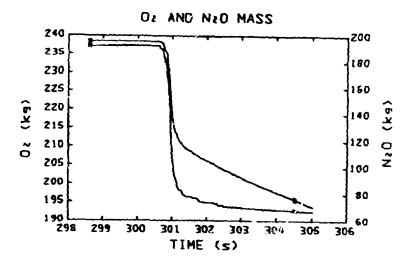


Fig. 3. Time history of O_2 and N_2O .

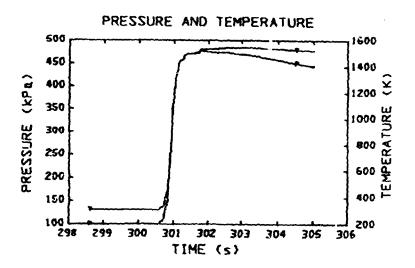


Fig. 5. Time history of the average pressure and temperature.